

Derivation of a time dependent Schrödinger equation as quantum mechanical Landau-Lifshitz-Bloch equation

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Abstract

The derivation of the time dependent Schrödinger equation with transversal and longitudinal relaxation, as the quantum mechanical analog of the classical Landau-Lifshitz-Bloch equation, has been described. Starting from the classical Landau-Lifshitz-Bloch equation the transition to quantum mechanics has been performed and the corresponding von-Neumann equation deduced. In a second step the time Schrödinger equation has been derived. Analytical proofs and computer simulations show the correctness and applicability of the derived Schrödinger equation.

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I. INTRODUCTION

The Landau-Lifshitz equation [1] respectively Landau-Lifshitz-Gilbert (LLG) equation [2] are the most prominent equations describing spin dynamics. These equations are intensively used to describe any kind of magnetization dynamics in ferromagnetic, antiferromagnetic or ferrimagnetic materials with diameters from a few Ångström (atomistic description) to a micrometer length scale (micromagnetism). The Landau-Lifshitz respectively LLG equation describes the motion of a magnetic moment under the influence of an effective field \mathbf{H}_{eff} which causes a precessional motion and an additional friction (transversal relaxation) which leads to a parallel alignment of the magnetic moment and the effective field. During the relaxation the length of the magnetic moment is conserved. However, there are situations, e.g. during ultrafast reversal processes [3] or the dynamics near the critical temperature T_C [4], where the description using the Landau-Lifshitz or LLG equation fails because the magnetization is not necessarily constant. This has been already pointed out by H. B. Callen in 1957 [5]: the general equation of motion of a ferromagnetic material has to be obtained by expanding the change of the magnetization \mathbf{M} in the three orthogonal vectors \mathbf{M} , $(\mathbf{M} \times \mathbf{H}_{\text{eff}})$, and $\mathbf{M} \times (\mathbf{M} \times \mathbf{H}_{\text{eff}})$.

$$\frac{d\mathbf{M}}{dt} = -\gamma\mathbf{M} \times \mathbf{H}_{\text{eff}} - \gamma\alpha_{\text{tr}}\mathbf{M} \times (\mathbf{M} \times \mathbf{H}_{\text{eff}}) - \gamma\alpha_l(\mathbf{M} \cdot \mathbf{H}_{\text{eff}})\mathbf{M}. \quad (1)$$

While $\gamma = g\mu_B/\hbar$ is the gyromagnetic ratio, α_{tr} and α_l are scalar functions of \mathbf{M} , and \mathbf{H}_{eff} . This equation is called Landau-Lifshitz-Bloch equation and without the last term ($\alpha_l = 0$) equal to the Landau-Lifshitz equation. In many cases this equation can be found for the corresponding magnetic moment $\mathbf{m} = \mathbf{M}V$, where V is the volume of the sample, or the normalized magnetic moment $\mathbf{S} = \mathbf{m}/|\mathbf{m}|$. Depending on the characteristics of the sample α_{tr} , α_l and \mathbf{H}_{eff} will be different. In general \mathbf{H}_{eff} is given by the negative gradient of the Hamiltonian \mathcal{H} with respect to the magnetization or magnetic moment e.g. $\mathbf{H}_{\text{eff}} = -\nabla_{\mathbf{M}}\mathcal{H}$, eventually modified by an additional stochastic noise term ξ describe the influence of temperature [6] and further modification to take into account that the magnetization and most of the material parameters itself like the anisotropy are temperature dependent [7].

As for the effective field we can find for α_{tr} and α_l different descriptions. The first proposal has been given by H. B. Callen [5]. The assumption there is that the dissipative process is

dominated by spin wave transitions from ($\mathbf{k} = 0$) to ($\mathbf{k} \neq 0$) where \mathbf{k} is the wave vector of the spin wave. Callen deduces for α_{tr} and α_1 (Eq. (36) in [5]):

$$\alpha_{\text{tr}} = \frac{1}{\gamma|\mathbf{H}_{\text{eff}}|} \left[\frac{1}{n_0} \frac{dn_0}{dt} + \frac{2\gamma\hbar}{|\mathbf{M}|} \frac{dn'}{dt} \right], \quad \alpha_1 = \frac{\hbar}{|\mathbf{M}|} \frac{dn'}{dt},$$

with $n' = \sum_{\mathbf{k} \neq 0} n_{\mathbf{k}}$ and $n_{\mathbf{k}} = a_{\mathbf{k}}^\dagger a_{\mathbf{k}}$, where $a_{\mathbf{k}}^\dagger$ and $a_{\mathbf{k}}$ are the Bose creation and annihilation operators.

R. S. Gekht et al. propose the following functions (see Eq. (3.4) in [8]):

$$\alpha_{\text{tr}} = \alpha, \quad \text{and} \quad \alpha_1 = \frac{2\alpha\gamma k_B T}{m},$$

with α the Gilbert damping constant, k_B the Boltzmann constant, T the temperature and $m = |\mathbf{m}|$. The assumption behind this proposal is the temperature dependence of the magnetization.

In 1990, the same idea following, D. A. Garanin et al. [9, 10] and T. Plefka [11, 12] proposed independently the following functions for α_{tr} and α_1 which can be found with just slightly changes in nearly all recent publications [4, 13–16] dealing with the Landau-Lifshitz-Bloch equation. Here in the writing of L. Xu and S. Zhang [15, 16]:

$$\alpha_{\text{tr}} = \frac{m_{\text{eq}}}{\gamma\tau_s m |\mathbf{H}_{\text{eff}}|}, \quad \alpha_1 = \frac{1}{\gamma\tau_s} \left[\frac{m}{\mathbf{m} \cdot \mathbf{H}_{\text{eff}}} - \frac{m_{\text{eq}}}{m |\mathbf{H}_{\text{eff}}|} \right].$$

$m_{\text{eq}} = |\mathbf{m}_{\text{eq}}|$ is the equilibrium magnetization and τ_s the spin relaxation time, similar to T_1 and T_2 in the case of the Bloch equation [17]. In the most cases the temperature dependence of m_{eq} is for simplicity reasons taken into account via a mean field theory.

In all these cases, even if the derivation starts with a quantum mechanical description, the authors end up with the (semi-) classical LLB equation [Eq. (1)] where \mathbf{M} is either the magnetization \mathbf{M} , the magnetic moment \mathbf{m} or \mathbf{S} or at least the spin expectation value $\langle \hat{\mathbf{S}} \rangle$ of the spin operator $\hat{\mathbf{S}}$. There are two reasons: The first reason can be seen in the Ehrenfest theorem [18], which says that the quantum mechanical expectation values behave classical. However, in the mean time it is known that the Ehrenfest theorem fails if the potential $V(x)$ is not linear: $\langle x^a \rangle \neq \langle x \rangle^a$ if $a \geq 2$. In the case of the Heisenberg model this means a classical behavior of the spin expectation values can be expected only if the terms of the Hamiltonian are linear in $\hat{\mathbf{S}}_n$, where n is the spin index [19]. This excludes especially crystalline anisotropies which are proportional to $\hat{\mathbf{S}}_n^2$.

The second reason is the fact that with the classical description larger system sizes as with a quantum mechanical description can be addressed. This can be explained with the fact that in the classical description the spins are local: every spin can be addressed separately and is affected by a local effective field. This makes it possible to simulate up to 10^6 spins [6]. In the quantum mechanical description we are dealing with wave functions describing all spins at the same time. The corresponding matrices are huge and actual it impossible to address more than 64 spins $S = 1/2$ in maximum using exact diagonalization [20]. The larger system sizes can be seen as an advantage. On the other hand with a classical description quantum effects get lost. The comparison between classical and quantum spin dynamics shows that a similar dynamics can be found only in some special cases:

1. in the classical limit ($S \rightarrow \infty$, $\hbar \rightarrow 0$, and $\hbar S \rightarrow 1$)
2. only linear terms in $\hat{\mathbf{S}}_n$, where n is the lattice site, in the Hamilton operator \hat{H} [19]
3. in the case of no entanglement [21], e.g. if the system is described by a product state [22]
4. $|\psi\rangle$ corresponds to a superposition of the basis states $|S, m_S\rangle = |S, \pm S\rangle$ only: $|\psi\rangle = \psi_{+S}|S, +S\rangle + \psi_{-S}|S, -S\rangle$.

The last scenario (point 4.) is the case for:

- (a) ferromagnetic spin waves: in this case $|\psi\rangle$ is approximately given by $|\psi\rangle \approx |S, \pm S\rangle$ [23, 24]
- (b) coherent states where $|\psi\rangle$ is given by $|\psi\rangle = U(\theta, \phi)|S, \pm S\rangle$, $U(\theta, \phi)$ is a unitary transformation describing a rotation with the rotation angles θ and ϕ [25]
- (c) a single spin with $S = 1/2$: in this case the wave function is always given by: $|\psi\rangle = \psi_{\uparrow}|\uparrow\rangle + \psi_{\downarrow}|\downarrow\rangle$ (Bloch sphere) [26]
- (d) a single spin with $S > 1/2$ if the only contribution to \hat{H} is a external field in direction of the quantization axis (in the most cases $\mathbf{B} = B_z \hat{\mathbf{z}}$). Perpendicular fields lead to quantum tunneling which can lead to states $|\psi\rangle = |S, m_S\rangle$, with $m_S \neq \pm S$ [21, 27–29].

As said before, the mentioned examples in the introduction using the Landau-Lifshitz-Bloch equation describing a classical or semiclassical spin dynamics which means they exclude

quantum effects like entanglement. The spin dynamics with or without entanglement is totally different. The spin expectation values $\langle \hat{\mathbf{S}} \rangle$ follow the same trajectories as the classical spin \mathbf{S} only if there is no entanglement [21]. This together with the possibility to find quantum tunneling in anisotropic spin systems [27–29] are the main differences between the quantum mechanical description which takes these effects into account and the classical or a semiclassical description which do not take into account these effects.

To take into account these quantum effects it is necessary to describe the system fully quantum mechanical and to calculate the spin expectation values at the end. The goal of this publication is to give a time dependent Schrödinger equation which enables us to address all quantum effects and at the same time to take into account transversal and longitudinal relaxation similar to the (semi-) classical description using the LLB equation [Eq. (1)].

The outline of the publication is the following: In Sec. II first the von Neumann equation will be introduced and after that the corresponding time dependent Schrödinger equation will be derived. The reason for this is the facts that the von Neumann equation is easier to understand and closer to the (semi-) classical description than the time dependent Schrödinger equation. However, the time dependent Schrödinger equation has a reduced numerical effort with respect to the von Neumann equation. For a Hilbert space of dimension N the number of components of the corresponding wave function $|\psi\rangle$ is N while the number of matrix components of the density operator matrix $\hat{\rho} = |\psi\rangle\langle\psi|$ is equal to N^2 [30]. In Sec. III the derived time dependent Schrödinger equation will be proved analytical. Here the Hamiltonian is chosen in such a way that we can expect a classical behavior of the spin expectation value $\langle \hat{\mathbf{S}} \rangle$. This gives us a direct proof of the correctness of our description. Sec. IV demonstrates the possibility to solve the derived time dependent Schrödinger equation under some more complex condition and the stability of the numerical calculation. The publication ends with a summary (Sec. V).

II. EQUATION OF MOTION

In a recently published manuscript [21] it has been shown that the following von Neumann equation:

$$\frac{d\hat{\rho}}{dt} = \frac{i}{\hbar}[\hat{\rho}, \hat{H}] - \frac{\alpha_{\text{tr}}}{\hbar}[\hat{\rho}, [\hat{\rho}, \hat{H}]] \quad (2)$$

and the corresponding self-consistent nonlinear Schrödinger equation:

$$i\hbar \frac{d}{dt}|\psi\rangle = \hat{H}|\psi\rangle - i\alpha_{\text{tr}} \left(\hat{H} - \langle \hat{H} \rangle \right) |\psi\rangle, \quad (3)$$

with $\langle \hat{H} \rangle = \langle \psi | \hat{H} | \psi \rangle$, and \hat{H} an arbitrary Hermitian Heisenberg Hamiltonian, can be used to describe the dynamics of quantum spin. In both equations the first term on the right hand side describes a precessional motion and the second term a transversal relaxation.

It has been further shown that in the case of a spin $\hat{\mathbf{S}}_n$, where n corresponds to the n th spin at lattice site \mathbf{r}_n , in an effective field \mathbf{B}_n the trajectories of the spin expectation value $\langle \hat{\mathbf{S}}_n \rangle = \langle \psi | \hat{\mathbf{S}}_n | \psi \rangle$, where the wave function $|\psi\rangle$ has been calculated with Eq. (3), are similar to the trajectories of the classical spin \mathbf{S}_n with dynamics described by the Landau-Lifshitz equation:

$$\frac{d\langle \hat{\mathbf{S}}_n \rangle}{dt} = \gamma \langle \hat{\mathbf{S}}_n \rangle \times \mathbf{B}_n - \gamma \alpha_{\text{tr}} \langle \hat{\mathbf{S}}_n \rangle \times \left(\langle \hat{\mathbf{S}}_n \rangle \times \mathbf{B}_n \right). \quad (4)$$

The only difference between both descriptions is the reversed sense of rotation of the precessional motion (first term: $\gamma \rightarrow -\gamma$).

However, there are two restrictions: Eq. (4) holds only if the Hamiltonian is linear in $\hat{\mathbf{S}}_n$ (point 2 in the list before). This means as long as we can write $\hat{H} = -\sum_n \mathbf{B}_n \cdot \hat{\mathbf{S}}_n$. Thereby, the effective field \mathbf{B}_n itself can be a function of the surrounding spins $\hat{\mathbf{S}}_m$, $m \neq n$, interacting with $\hat{\mathbf{S}}_n$. Higher order contributions as uniaxial anisotropies or the biquadratic exchange (both quadratic in $\hat{\mathbf{S}}_n$), will lead to additional terms of the order \hbar , which disappear in the classical limit $\hbar \rightarrow 0$ [19].

The second restriction is related to the entanglement. While Eq. (4) looks similar to the classical Landau-Lifshitz equation the trajectories of spin expectation values $\langle \hat{\mathbf{S}}_n \rangle$ and the corresponding classical spin \mathbf{S}_n are not necessary equal. The trajectories differ if entanglement plays a role. While in the (semi-) classical description the length of the spin $|\mathbf{S}_n|$ only changes if there is an additional longitudinal relaxation as in the Landau-Lifshitz-Bloch equation the value $|\langle \hat{\mathbf{S}}_n \rangle|$ can also change if there is no longitudinal relaxation in the equation of motion as in Eq. (4). Indeed, $|\langle \hat{\mathbf{S}}_n \rangle|$ is a direct indicator for entanglement: $|\langle \hat{\mathbf{S}}_n \rangle| = \hbar S$ can be expected only if the system shows no entanglement [21]. In all the other cases we have $|\langle \hat{\mathbf{S}}_n \rangle| < \hbar S$. In a case of maximal entanglement as in the case of singlet state $|\text{singlet}\rangle = (|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle)/\sqrt{2}$ we have $|\langle \hat{\mathbf{S}}_n \rangle| = 0$ [22].

To take the entanglement into account it is mandatory necessary that we solve the time dependent Schrödinger Eq. (3) or the corresponding von Neumann Eq. (2) and in a second

step calculate the spin expectation values $\langle \hat{\mathbf{S}}_n \rangle$. Eq. (4) does not take into account the change of $|\langle \hat{\mathbf{S}}_n \rangle|$ and therefore entanglement. This means the description where we solve Eq. (4) to calculate the trajectories of $\langle \hat{\mathbf{S}}_n \rangle$ has to be considered as semiclassical. The resulting trajectories in this case will be the same as for the classical Landau-Lifshitz equation but not necessarily the same as in the case of the quantum mechanical description. The same is true for the Landau-Lifshitz-Bloch equation and the corresponding Schrödinger equation which we will derive in the following.

The first step will be to derive the corresponding von Neumann equation. As going from the Landau-Lifshitz equation [Eq. (4)] to the Landau-Lifshitz-Bloch equation [Eq. (1)] we have to add to the von Neumann equation [Eq. (2)] an additional longitudinal relaxation term which can be derived from the longitudinal relaxation term of the classical Landau-Lifshitz-Bloch equation: $\gamma\alpha_1 (\mathbf{S} \cdot \mathbf{B}) \mathbf{S}$ (we assume $\mathbf{H}_{\text{eff}} = \mathbf{B}$) in the following way: we replace the classical spin \mathbf{S} by the expectation value $\langle \hat{\mathbf{S}} \rangle$ and correct the dimension. \mathbf{S} is assumed to be dimensionless, but $\langle \hat{\mathbf{S}} \rangle$ has the dimension of \hbar . We correct the dimension by an additional $1/\hbar$ to keep α_1 dimensionless:

$$- \gamma\alpha_1 (\mathbf{S} \cdot \mathbf{B}) \mathbf{S} \rightarrow -2\frac{\alpha_1}{\hbar} \left(\frac{g\mu_B}{\hbar} \mathbf{B} \cdot \langle \hat{\mathbf{S}} \rangle \right) \langle \hat{\mathbf{S}} \rangle = 2\frac{\alpha_1}{\hbar} \langle \hat{H} \rangle \langle \hat{\mathbf{S}} \rangle .$$

The same dimension problem also appears for α_{tr} . We have to correct the dimension there too. The additional factor 2 is needed to guarantee later a symmetric decoupling during the derivation of the time-dependent Schrödinger equation. Furthermore, it can be shown that this factor is needed to get the correct spin length (see supplementary material [31]).

The next step is to write $\langle \hat{H} \rangle \langle \hat{\mathbf{S}} \rangle$ as:

$$\langle \hat{H} \rangle \langle \hat{\mathbf{S}} \rangle = \text{Tr}(\hat{\rho} \hat{H}) \text{Tr}(\hat{\rho} \hat{\mathbf{S}}) = \text{Tr}(\hat{\rho} \hat{H} \hat{\rho} \hat{\mathbf{S}}) \quad (5)$$

[21]. The relaxation term in terms of the density operator $\hat{\rho}$ appears if we skip Tr and $\hat{\mathbf{S}}$ in Eq. (5): $\text{Tr}(\hat{\rho} \hat{H} \hat{\rho} \hat{\mathbf{S}}) \rightarrow \hat{\rho} \hat{H} \hat{\rho}$. Adding the resulting expression to Eq. (2) leads to the von Neumann equation containing transversal and longitudinal relaxation:

$$\frac{d\hat{\rho}}{dt} = \frac{i}{\hbar} [\hat{\rho}, \hat{H}] - \frac{\alpha_{\text{tr}}}{\hbar} [\hat{\rho}, [\hat{\rho}, \hat{H}]] + 2\frac{\alpha_1}{\hbar} (\hat{\rho} \hat{H}) \hat{\rho} . \quad (6)$$

To derive the corresponding Schrödinger equation we use the definition of the density operator in the case of a pure state:

$$\hat{\rho} = |\psi\rangle \langle \psi| . \quad (7)$$

Inserting $\hat{\rho}$ in Eq. (6) we find after some algebra the following two differential equations:

$$\begin{aligned} \frac{d|\psi\rangle}{dt}\langle\psi| &= \left[-\frac{i}{\hbar}\hat{H}|\psi\rangle - \frac{\alpha_{\text{tr}}}{\hbar}(\hat{H} - \langle\hat{H}\rangle)|\psi\rangle \right] \langle\psi| \\ &+ \frac{\alpha_1}{\hbar}|\psi\rangle\langle\hat{H}|\langle\psi| \end{aligned} \quad (8)$$

$$\begin{aligned} |\psi\rangle\frac{d\langle\psi|}{dt} &= |\psi\rangle\left[\langle\psi|\hat{H}\frac{i}{\hbar} - \langle\psi|(\hat{H} - \langle\hat{H}\rangle)\frac{\alpha_{\text{tr}}}{\hbar} \right] \\ &+ \frac{\alpha_1}{\hbar}|\psi\rangle\langle\hat{H}|\langle\psi|. \end{aligned} \quad (9)$$

After multiplying Eq. (8) with $|\psi\rangle$ from the right and dividing both sides by $\langle\psi|\psi\rangle$ we find the Schrödinger equation:

$$i\hbar\frac{d}{dt}|\psi\rangle = (\hat{H} - i\alpha_{\text{tr}}[\hat{H} - \langle\hat{H}\rangle] + i\alpha_1\langle\hat{H}\rangle)|\psi\rangle. \quad (10)$$

The three terms on the right hand side are the same as in the case of the classical Landau-Lifshitz-Bloch equation. The first term describes an undamped precession. The second term provides a transversal and the last term a longitudinal relaxation. In the case of the Landau-Lifshitz-Bloch equation the transversal and longitudinal relaxation act separate, which means independent. However, a careful analysis shows that this is not the case for the recently proposed Schrödinger equation. This equation has a problem if we assume a spin oriented parallel to an external field $\mathbf{B} = B_z\hat{\mathbf{z}}$. In this case only the longitudinal relaxation should contribute and lead to a change the length of the spin. A transversal relaxation should taking place. However, it can be shown that in this simple scenario both relaxation terms influence the change of the spin length. The influence of the transversal relaxation term can be seen with $\hat{H} = -g\mu_B B_z \hat{S}_z/\hbar = -b_z \hat{\sigma}_z$ and $|\psi(t)\rangle = \psi_\uparrow(t)|\uparrow\rangle = \psi_\uparrow(t)(1,0)^T$ by:

$$-i\alpha_{\text{tr}}[\hat{H} - \langle\hat{H}\rangle]|\psi\rangle = ib_z\alpha_{\text{tr}}\psi_\uparrow(t)[1 - |\psi_\uparrow|^2] \begin{pmatrix} 1 \\ 0 \end{pmatrix}.$$

This equation only becomes zero if $|\psi_\uparrow|^2 = \langle\psi|\psi\rangle = 1$, which means in the case of a normalized wave function. This is surely the case for $\alpha_1 = 0$ (no longitudinal relaxation), where we deal with normalized wave functions [21]. However, in the cases $\alpha_1 > 0$ we have a decrease or increase of the norm of the wave function $n = \langle\psi|\psi\rangle$ due to the longitudinal relaxation. In this case we have $0 \leq \langle\psi|\psi\rangle \leq 1$ and therefore a contribution of the transversal relaxation. This means we need a modification to fix this problem:

$$i\hbar\frac{d}{dt}|\psi\rangle = (\hat{H} - i\alpha_{\text{tr}}[\langle\psi|\psi\rangle\hat{H} - \langle\hat{H}\rangle] + i\alpha_1\langle\hat{H}\rangle)|\psi\rangle. \quad (11)$$

The modification is to add $\langle\psi|\psi\rangle$ into the transversal relaxation term which leads to $-i\alpha_{\text{tr}}[\langle\psi|\psi\rangle\hat{H} - \langle\hat{H}\rangle]|\psi\rangle = 0$ in any case. With this modification the transversal relaxation does not contribute in this constellation and we can expect the correct results.

III. ANALYTICAL PROOF

It is easy to show that the conjugate transposed equation corresponding to Eq. (11) is given by:

$$-i\hbar\frac{d}{dt}\langle\psi| = \langle\psi|(\hat{H} + i\alpha_{\text{tr}}[\hat{H}\langle\psi|\psi\rangle - \langle\hat{H}\rangle] - i\alpha_1\langle\hat{H}\rangle) . \quad (12)$$

Next, we are looking for the time development of a single spin in an external field \mathbf{B} described by:

$$\mathbf{m} = \frac{\langle\hat{\mathbf{S}}\rangle}{\hbar S} , \quad (13)$$

with $\langle\hat{\mathbf{S}}\rangle = \langle\psi|\hat{\mathbf{S}}|\psi\rangle$. Furthermore, we assume $S = 1/2$ which means $\hat{\mathbf{S}} = \frac{\hbar}{2}\hat{\sigma}$ and

$$\mathbf{m} = \langle\psi|\hat{\sigma}|\psi\rangle . \quad (14)$$

Therefore, the time derivative of $\mathbf{m}(t)$ is given by:

$$\frac{d\mathbf{m}}{dt} = \langle\dot{\psi}|\hat{\sigma}|\psi\rangle + \langle\psi|\hat{\sigma}|\dot{\psi}\rangle , \quad (15)$$

where $|\dot{\psi}\rangle = \frac{d}{dt}|\psi\rangle$ and $\langle\dot{\psi}| = \frac{d}{dt}\langle\psi|$ are represented by the time dependent Schrödinger equation (11) and the corresponding conjugate transposed equation (12).

It is more convenient to look for one component of \mathbf{m} , e.g z -component m_z . The time development of m_z is given by:

$$\frac{dm_z}{dt} = \langle\dot{\psi}|\hat{\sigma}_z|\psi\rangle + \langle\psi|\hat{\sigma}_z|\dot{\psi}\rangle . \quad (16)$$

Due to the assumption $S = 1/2$ the Hamilton operator of a single spin in an external field is given by:

$$\hat{H} = -\frac{g\mu_B}{\hbar}\mathbf{B} \cdot \hat{\mathbf{S}} = -\frac{g\mu_B}{2}\mathbf{B} \cdot \hat{\sigma} , \quad (17)$$

and $\langle\hat{H}\rangle$ as:

$$\langle\hat{H}\rangle = -\frac{g\mu_B}{2}\mathbf{B} \cdot \mathbf{m} . \quad (18)$$

Inserting the equations of motions Eq. (11) and (12), together with Eq. (17) and (18) in Eq. (16) we get after some algebra:

$$\begin{aligned}
\frac{dm_z}{dt} = & -\frac{ig\mu_B}{2\hbar} (B_x \langle \psi | [\hat{\sigma}_x, \hat{\sigma}_z] | \psi \rangle + B_y \langle \psi | [\hat{\sigma}_y, \hat{\sigma}_z] | \psi \rangle) \\
& + \frac{g\mu_B \alpha_{\text{tr}}}{2\hbar} \langle \psi | \psi \rangle B_x \langle \psi | \{ \hat{\sigma}_x, \hat{\sigma}_z \} | \psi \rangle \\
& + \frac{g\mu_B \alpha_{\text{tr}}}{2\hbar} \langle \psi | \psi \rangle B_y \langle \psi | \{ \hat{\sigma}_y, \hat{\sigma}_z \} | \psi \rangle \\
& + \frac{g\mu_B \alpha_{\text{tr}}}{\hbar} B_z [\langle \psi | \psi \rangle]^2 - \frac{g\mu_B (\alpha_{\text{tr}} + \alpha_1)}{\hbar} (\mathbf{B} \cdot \mathbf{m}) m_z
\end{aligned} \tag{19}$$

Here, we have used the definition of $m_z = \langle \psi | \hat{\sigma}_z | \psi \rangle$ and $\hat{\sigma}_z \hat{\sigma}_z = \hat{\mathbf{1}}$, where $\hat{\mathbf{1}}$ is the identity matrix. The same is true for the other Pauli matrices: $\hat{\sigma}_x \hat{\sigma}_x = \hat{\sigma}_y \hat{\sigma}_y = \hat{\mathbf{1}}$. In Eq. (19) the $[\hat{\sigma}_\alpha, \hat{\sigma}_\beta] = \hat{\sigma}_\alpha \hat{\sigma}_\beta - \hat{\sigma}_\beta \hat{\sigma}_\alpha$ are commutators while the $\{\hat{\sigma}_\alpha, \hat{\sigma}_\beta\} = \hat{\sigma}_\alpha \hat{\sigma}_\beta + \hat{\sigma}_\beta \hat{\sigma}_\alpha$ are anticommutators. Independent of S the commutators are given by $[\hat{\sigma}_\alpha, \hat{\sigma}_\beta] = 2i\epsilon_{\alpha,\beta,\gamma} \hat{\sigma}_\gamma$, where $\epsilon_{\alpha,\beta,\gamma}$ is the Levi-Civita tensor. With changing S , only the Pauli matrices change. This is not the case for the anticommutators $\{\hat{S}_\alpha, \hat{S}_\beta\}$. They are changing with S . In the case of $S = 1/2$ the anticommutators are given by: $\{\hat{\sigma}_\alpha, \hat{\sigma}_\beta\} = 2\delta_{\alpha,\beta} \hat{\mathbf{1}}$. However, this is not the case for $S > 1/2$. The general anticommutator relations for the spin operator \hat{S}_α are given by: $\{\hat{S}_\alpha, \hat{S}_\beta\} = 4/N \delta_{\alpha\beta} \hat{\mathbf{1}} + 2g_{\alpha\beta\gamma} \hat{S}_\gamma$, where $g_{\alpha\beta\gamma}$ is the completely symmetric tensor of the Lie algebra $su(N)$, and N the number of quantum level [32]. For $S = 1/2$ we have $N = 2$, and $g_{\alpha\beta\gamma} = 0$.

After working out the commutators and anticommutators we find with $\gamma = g\mu_B/\hbar$:

$$\begin{aligned}
\frac{dm_z}{dt} = & \gamma [m_x B_y - m_y B_x] - \alpha_1 \gamma (\mathbf{B} \cdot \mathbf{m}) m_z \\
& - \alpha_{\text{tr}} \gamma [m_z (\mathbf{B} \cdot \mathbf{m}) - B_z \underbrace{(\tilde{\mathbf{m}} \cdot \tilde{\mathbf{m}})}_{=1} [\langle \psi | \psi \rangle]^2] .
\end{aligned} \tag{20}$$

Here, the definitions for $m_x = \langle \psi | \hat{\sigma}_x | \psi \rangle$ and $m_y = \langle \psi | \hat{\sigma}_y | \psi \rangle$ have been used and the assumption that $\tilde{\mathbf{m}} = \langle \psi | \hat{\sigma} | \psi \rangle / \langle \psi | \psi \rangle$ is normalized: $\tilde{\mathbf{m}} \cdot \tilde{\mathbf{m}} = \tilde{\mathbf{m}}^2 = 1$.

The last equation can be written in a more compact form using the vector triple product identity and $\mathbf{m} = \tilde{\mathbf{m}} \langle \psi | \psi \rangle$:

$$\begin{aligned}
\frac{dm_z}{dt} = & \gamma [\mathbf{m} \times \mathbf{B}]_z - \alpha_{\text{tr}} \gamma [\mathbf{m} \times (\mathbf{m} \times \mathbf{B})]_z \\
& - \alpha_1 \gamma (\mathbf{B} \cdot \mathbf{m}) m_z .
\end{aligned} \tag{21}$$

The equations for m_x and m_y can be derived in a similar way and therefore, we finally get:

$$\frac{d\mathbf{m}}{dt} = \gamma \mathbf{m} \times \mathbf{B} - \alpha_{\text{tr}} \gamma \mathbf{m} \times (\mathbf{m} \times \mathbf{B}) - \alpha_1 \gamma (\mathbf{B} \cdot \mathbf{m}) \mathbf{m} . \quad (22)$$

This equation is identical to the classical Landau-Lifshitz-Bloch equation, if we ignore the sign problem of the precessional motion ($\gamma \rightarrow -\gamma$).

IV. NUMERICAL EXAMPLES

In the last section III the proposal has been proved analytical for the case of a single spin with $S = 1/2$. We have seen that in this case the Schrödinger equation [Eq. (11)] leads to an equation for the expectation values $\mathbf{m} = \langle \psi | \hat{\mathbf{S}} | \psi \rangle$ which is similar to the classical Landau-Lifshitz-Bloch equation. To strengthen this statement we present in this section computer simulations to show the correctness and the possibilities of the given description.

For the computer simulations we have solved the time dependent Schrödinger equation [Eq. (10)] numerical for different scenarios. In the following we set $\hbar = 1$ which means that the time will be in natural units: $t_{\text{sim.}} = \hbar t_{\text{real.}}$. Under the assumption that we have energies in units of electronvolts the time scales of the simulations are in the femtosecond regime.

In the first scenario we assume a starting configuration of one spin with spin quantum number $S = 1/2$ oriented in $+x$ -direction: $|\psi\rangle_{\text{init}} = (|\uparrow\rangle + |\downarrow\rangle)/\sqrt{2}$ and a length $|\langle \hat{\mathbf{S}} \rangle|/\hbar S = 1$. Furthermore, an external field in $+z$ -direction: $\mathbf{B} = B_z \hat{\mathbf{z}}$. The scenario has been chosen in such a way that, following the description (point 4c), we can expect a behavior of the spin expectation value $\langle \hat{\mathbf{S}} \rangle$ similar to the dynamics of a classical spin \mathbf{S} (except the different sense of rotation). The Hamiltonian of this scenario is given by:

$$\hat{H} = -\frac{g\mu_B}{\hbar} B_z \hat{S}_z . \quad (23)$$

Due to the relaxation terms the spin will relax into the direction of the external field and shrink to the equilibrium length $|\langle \hat{\mathbf{S}} \rangle_{\text{eq}}|/\hbar S = 0.7$. For α_{tr} and α_1 we use the definitions:

$$\alpha_{\text{tr}} = \alpha_{\text{tr}}^0 \quad \text{and} \quad \alpha_1 = \alpha_1^0 \cdot \left(\frac{|\langle \hat{\mathbf{S}} \rangle| - |\langle \hat{\mathbf{S}} \rangle_{\text{eq}}|}{\hbar S} \right) , \quad (24)$$

with $\alpha_{\text{tr}}^0 = 0.02$ and $\alpha_1^0 = 0.04$.

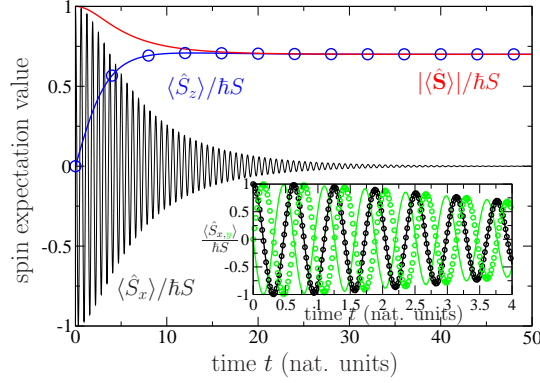


FIG. 1: (color online) Relaxation of a single spin ($S = 1/2$) with initial orientation in $+x$ -direction and length $|\langle \hat{\mathbf{S}} \rangle| / \hbar S = 1$ in an external field in $+z$ -direction and a final spin length $|\langle \hat{\mathbf{S}} \rangle| / \hbar S = 0.7$. The solid lines correspond to the quantum mechanical expectation values $\langle \hat{\mathbf{S}} \rangle$ and the circles the classical trajectories \mathbf{S} . (Simulation parameter: $\hbar = 1$, $\gamma B_z = 10$, $\alpha_{\text{tr}}^0 = 0.02$, $\alpha_1^0 = 0.04$)

Fig. 1 presents the relaxation process of a quantum spin ($S = 1/2$) compared with a classical spin ($S = \infty$). In the later case we have solved the Landau-Lifshitz equation. In both cases the spin has been normalized and in the case of the classical spin we have used the double field value: $2B_z$ instead of B_z . The doubling of the field value is necessary to make the quantum mechanical and the classical trajectories comparable. The reason for that are the different Zeeman energies qm: $\hat{H} = -g\mu_B \mathbf{B} \cdot \hat{\sigma}/2$ [see Eq. (17)] and cl.: $H = -g\mu_B \mathbf{B} \cdot \mathbf{S}$. Fig. 1 clearly shows that in this case we find a perfect agreement between the classical trajectories \mathbf{S} and quantum mechanical expectation values $\langle \hat{\mathbf{S}} \rangle$. The only difference appears for $\langle \hat{S}_x \rangle$. Here, we see a phase difference of 180° coming from the different rotation senses of the precession in the classical and quantum spin dynamics ($\gamma \rightarrow -\gamma$ problem). However, the amplitude and frequency are the same.

To show the reliability of the given description we have performed more complex simulations. The next scenario has an initial configuration with a single spin with $S = 1/2$ oriented in $+z$ -direction with $|\langle \hat{\mathbf{S}} \rangle| / \hbar S = 1$. As before we assume an external magnetic field in $+z$ direction plus an additional Gaussian field pulse:

$$\mathbf{B}_x(t) = B_0^x e^{-\frac{1}{2} \left(\frac{t-t_0}{T_W} \right)^2} \hat{\mathbf{x}} \quad (25)$$

in x -direction to excite the spin. Therefore, the Hamilton operator of this scenario is:

$$\hat{H} = -\frac{g\mu_B}{\hbar} \left(B_x(t) \hat{S}_x + B_z \hat{S}_z \right), \quad (26)$$

with $B_x(t)$ given by Eq. (25).

For the damping parameters α_{tr} and α_l the definitions given by L. Xu and S. Zhang [15, 16] have been used. Within their publication L. Xu and S. Zhang have proposed the following von Neumann equation:

$$\frac{d\hat{\rho}}{dt} = \frac{i}{\hbar} [\hat{\rho}, \hat{H}] - \frac{\hat{\rho} - \hat{\rho}_{\text{eq}}}{\tau_S}, \quad (27)$$

which becomes with $\hat{H} = -\frac{g\mu_B}{\hbar} \mathbf{B} \cdot \hat{\mathbf{S}}$ in the classical limit:

$$\frac{d\mathbf{m}}{dt} = \gamma \mathbf{m} \times \mathbf{B} - \frac{\mathbf{m} - \mathbf{m}_{\text{eq}}}{\tau_S}. \quad (28)$$

\mathbf{m}_{eq} is the equilibrium magnetization and τ_S is a constant describing the strength of the relaxation.

L. Xu and S. Zhang have shown that with the identity: $\mathbf{B} = m^{-2} [(\mathbf{m} \cdot \mathbf{B}) \mathbf{m} - \mathbf{m} \times (\mathbf{m} \times \mathbf{B})]$ this equation becomes:

$$\frac{d\mathbf{m}}{dt} = \gamma \mathbf{m} \times \mathbf{B} - \gamma \alpha_{\text{tr}} \mathbf{m} \times (\mathbf{m} \times \mathbf{B}) - \gamma \alpha_l (\mathbf{m} \cdot \mathbf{B}) \mathbf{m}, \quad (29)$$

which is identical to the Landau-Lifshitz-Bloch Eq. (1).

The damping parameters are given by:

$$\alpha_{\text{tr}} = \frac{m_{\text{eq}}}{\gamma \tau_S m B}, \quad (30)$$

as well as

$$\alpha_l = \frac{1}{\gamma \tau_S} \left[\frac{m}{\mathbf{m} \cdot \mathbf{B}} - \frac{m_{\text{eq}}}{m B} \right]. \quad (31)$$

To make Eq. (29) more general we replace τ_S in Eq. (30) by τ_{tr} and in Eq. (31) by τ_l . τ_{tr} and τ_l are similar to τ_S constants. Furthermore, we replace the classical \mathbf{m} , $m = |\mathbf{m}|$, and m_{eq} by their quantum mechanical counterparts $\langle \hat{\mathbf{S}} \rangle$, $|\langle \hat{\mathbf{S}} \rangle|$, and $|\langle \hat{\mathbf{S}} \rangle_{\text{eq}}|$. The results are the following two damping parameters:

$$\alpha_{\text{tr}} = \frac{\hbar}{g\mu_B \tau_{\text{tr}}} \frac{|\langle \hat{\mathbf{S}} \rangle_{\text{eq}}|}{|\langle \hat{\mathbf{S}} \rangle| B}, \quad (32)$$

and

$$\alpha_l = \frac{\hbar}{g\mu_B \tau_l} \left[\frac{|\langle \hat{\mathbf{S}} \rangle|}{\langle \hat{\mathbf{S}} \rangle \cdot \mathbf{B}} - \frac{|\langle \hat{\mathbf{S}} \rangle_{\text{eq}}|}{|\langle \hat{\mathbf{S}} \rangle| B} \right]. \quad (33)$$

Fig. 2 shows the x and z components of the spin expectation value $\langle \hat{\mathbf{S}} \rangle$ as well as the length $|\langle \hat{\mathbf{S}} \rangle|$ of a single spin in an external field oriented in $+z$ direction as function of time.

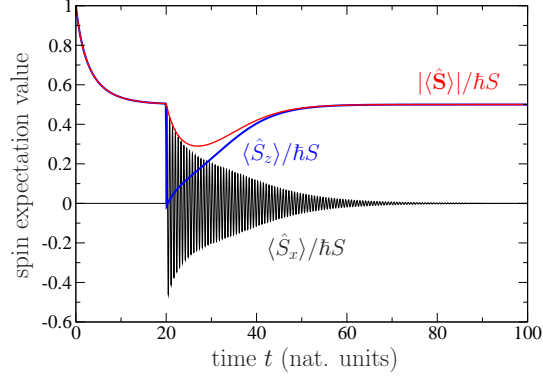


FIG. 2: (color online) Longitudinal relaxation and combined transversal + longitudinal relaxation after a Gaussian field pulse at $t = 20$ for a quantum spin with $S = 1/2$, initial and final spin length $|\langle\hat{\mathbf{S}}\rangle_{\text{init}}|/\hbar S = 1$ respectively $|\langle\hat{\mathbf{S}}\rangle_{\text{final}}|/\hbar S = 0.5$. (Simulation parameter: $\hbar = 1$, $\gamma B_z = 10$, $\gamma B_0^x = 46.54$, $(\gamma\tau_{\text{tr}})^{-1} = 0.1$, $(\gamma\tau_1)^{-1} = 0.2$, $t_0 = 20$, $T_W = 0.02$)

The initial spin is oriented parallel to the external field and has a length of $|\langle\hat{\mathbf{S}}\rangle| = 1$. The equilibrium length $|\langle\hat{\mathbf{S}}\rangle_{\text{eq}}|$ has been chosen as $|\langle\hat{\mathbf{S}}\rangle_{\text{eq}}| = 0.5$. Therefore, and due to the fact that there is only the external field in $+z$ direction only the longitudinal relaxation contributes to the dynamics. Fig. 2 clearly shows that the z component of $\langle\hat{\mathbf{S}}\rangle$ decays exponentially with the time until it reaches the equilibrium length $|\langle\hat{\mathbf{S}}\rangle_{\text{eq}}| = 0.5$. After reaching the equilibrium a Gaussian field pulse has been applied bringing the z component $\langle\hat{S}_z\rangle$ close to zero. After the field pulse the spin relaxes back to equilibrium, but this time all three terms: precession, transverse and longitudinal relaxation contribute to the dynamics.

The last example shall demonstrate that the given description is not restricted to a single spin. In the following we assume two spins $S = 1/2$ antiferromagnetically exchange coupled and where the first spin can be manipulated by an external field:

$$\hat{H} = J \frac{\hat{\mathbf{S}}_1 \cdot \hat{\mathbf{S}}_2}{\hbar^2} - \frac{g\mu_B}{\hbar} B_1^z(t) \hat{S}_1^z \quad (34)$$

The first term describes the antiferromagnetic exchange coupling with $J > 0$. The second term describes the coupling between first spin and an external field which is time dependent. This external field can be seen as a rough description of an electric current of a spin-polarized scanning tunneling microscope [33] or as an approximate description of the coupling to a magnetic island as described in [34]. In both cases we assume that we can switch the field and therefore the influence on and off.

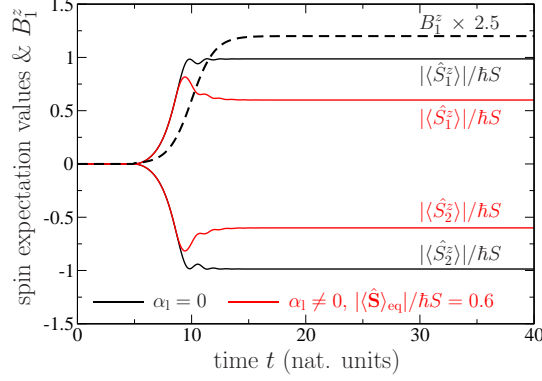


FIG. 3: (color online) Normalized spin expectation values $\langle \hat{S}_n^z \rangle / \hbar S$, $n \in \{1, 2\}$ and external field B_1^z (2.5 times smaller than in real) as function of time t . The different colors correspond to the different assumptions of α_1 : $\alpha_1^0 = 0$ and $\alpha_1^0 = 1.0$ together with $|\langle \hat{\mathbf{S}} \rangle_{\text{eq}}| / \hbar S = 0.6$. (Simulation parameter: $\hbar = 1$, $J/\hbar^2 = 1$, $\gamma B_0^z = 3$, $\kappa = 0.5$, $t_0 = 10$, $\alpha_{\text{tr}}^0 = 0.2$, $\alpha_1^0 = 1.0$)

In the case of a magnetic island it means that we e.g. increase the temperature above the Curie temperature T_C to switch the field off and let the island cool down to switch it on again. We further assume that we start with a zero external field which increases with the time:

$$B_1^z(t) = B_0^z \tanh[\kappa(t - t_0)] + B_0^z, \quad (35)$$

where $B_0^z = 3.0$ and $\kappa = 0.5$ are constants describing the maximum field strength and the inclination with the time and $t_0 = 10$.

As long as the field is switched off ($B_1^z(t) = 0$) the spins are in the ground state configuration which means in this case the singlet state:

$$|\psi\rangle = \frac{1}{\sqrt{2}} (|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle). \quad (36)$$

With $B_1^z(t) > 0$ the first spin becomes stabilized and we find as final state:

$$|\psi\rangle = \sqrt{|\langle \hat{\mathbf{S}} \rangle_{\text{eq}}|} |\uparrow\downarrow\rangle. \quad (37)$$

$|\langle \hat{\mathbf{S}} \rangle_{\text{eq}}|$ has been determined at the beginning of the calculation as part of the definition of α_1 [see Eq. (24) or (33)]. During the simulation for the damping terms the definition Eq. (24) with $\alpha_{\text{tr}}^0 = 0.2$ and $\alpha_1^0 = 1.0$ has been used. Fig. 3 shows the normalized spin expectation values $\langle \hat{S}_n^z \rangle / \hbar S$, $n \in \{1, 2\}$ of the first and second spin, as well as the time dependent external field $B_1^z(t)$. The zero expectation values $\langle \hat{S}_1^z \rangle / \hbar S = \langle \hat{S}_2^z \rangle / \hbar S = 0$ correspond to the initial

singulet state [Eq. (36)] and $B_1^z = 0$. With $B_1^z > 0$ the expectation values $\langle \hat{S}_n^z \rangle / \hbar S$ increase / decrease to their final values $\pm |\langle \hat{\mathbf{S}} \rangle_{\text{eq}}|$ corresponding to $|\psi\rangle_{\text{final}}$ given by Eq. (37) (classical Néel state). The other expectation values $\langle \hat{S}_n^x \rangle / \hbar S$ and $\langle \hat{S}_n^y \rangle / \hbar S$ are zero for all times.

V. SUMMARY

Within this publication the way how to derive the time dependent Schrödinger equation which can be seen as the quantum mechanical analog to the classical Landau-Lifshitz-Bloch equation has been demonstrated. The starting point is the Landau-Lifshitz-Bloch equation itself. From this equation the corresponding von Neumann equation can be deduced. And the von Neumann equation is the starting point to derive the corresponding time dependent Schrödinger equation. Therefore, with von Neumann equation and the time dependent Schrödinger equation we have two equations which can be seen as quantum mechanical analogs to the classical Landau-Lifshitz-Bloch equation. This opens the opportunity to extend the spin dynamics with transversal and longitudinal relaxation to the quantum regime and to compare the classical with the quantum spin dynamics.

The correctness of the time dependent Schrödinger equation and therefore indirectly also of the von Neumann equation has been proved analytical and with computer simulations. It has been shown that derived Schrödinger equation can lead to the same dynamics as the classical Landau-Lifshitz-Bloch equation. However, in the most scenarios we have to expect a different dynamics due to quantum effects. With the given description we have a general description of the quantum spin dynamics with transversal and longitudinal relaxation which is simple to understand. However, the given description will not give an insight to the microscopic processes of the damping. The energy gain and loss is introduced by phenomenological damping functions. This is the same in the case of the classical description. This can be seen as a disadvantage. On the other hand the Landau-Lifshitz-Gilbert as well as the Bloch equation became successful due to their simplicities and the fact that it is not necessary to know the underlying damping mechanism. In the given description this is the same and can be seen as an advantage. Furthermore, due to the fact that the damping parameters are no longer constant, but functions the given description is quite general. E.g. using the description of coherent states for $|\psi\rangle$ [25, 35] together with the definition for α_{tr} and α_1 in [36] reproduces the Landau-Lifshitz-Bloch equation which has been successful used

to describe ultrafast magnetization dynamics measurements.

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**Supplementary Material: Derivation of a time dependent
Schrödinger equation as quantum mechanical
Landau-Lifshitz-Bloch equation**

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I. INTRODUCTION

The following sections gives an overview about the quantum spin dynamics using the von Neumann equation with additional transversal and longitudinal relaxation terms as equation of motion to describe quantum spin dynamics. The work is still in progress and maybe not all the derivations and descriptions are not hundred percent correct. However, the idea of this supplementary is to give an introduction in this topic and to present some concepts.

II. DERIVATION OF THE VON NEUMANN EQ. WHICH CAN BE SEEN AS THE QUANTUM MECHANICAL ANALOG TO THE LANDAU-LIFSHITZ EQUATION

Starting point is the non-Hermitian Hamiltonian operator

$$\hat{\mathcal{H}} = \hat{H} - i\alpha\hat{H} , \quad (1)$$

with \hat{H} a Hermitian Heisenberg model Hamilton operator and $\alpha \in \mathbb{R}_0^+$ a constant. Such an Hamiltonian leads to energy dissipation and at the same time does not conserve the norm of the wave function:

$$n = \langle \psi(t) | \psi(t) \rangle = \langle \psi_0 | e^{-2\alpha\hat{H}t} | \psi_0 \rangle = e^{-2\alpha\langle\hat{H}\rangle t} . \quad (2)$$

However, the norm can be conserved by replacing $i\alpha\hat{H}$ by $i\alpha(\hat{H} - \langle\hat{H}\rangle)$

$$|\psi(t)\rangle = e^{-i\hat{H}t} e^{-\alpha\hat{H}t} e^{\alpha\langle\hat{H}\rangle t} |\psi_0\rangle . \quad (3)$$

In this case the norm keeps constant $n = 1$.

The corresponding Schrödinger equation is given by:

$$i\hbar \frac{d}{dt} |\psi(t)\rangle = (\hat{H} - i\alpha[\hat{H} - \langle\hat{H}\rangle]) |\psi(t)\rangle . \quad (4)$$

Eq. (4) can be easily written as:

$$i\hbar \frac{d}{dt} |\psi\rangle = \left(\hat{H} - i\alpha \left[\hat{H}, |\psi\rangle\langle\psi| \right] \right) |\psi\rangle . \quad (5)$$

The corresponding conjugate transposed equation is given by:

$$-i\hbar \frac{d}{dt} \langle\psi| = \langle\psi| \left(\hat{H} + i\alpha \left[|\psi\rangle\langle\psi|, \hat{H} \right] \right) . \quad (6)$$

With these equations, we are able to construct a von Neumann equation:

$$\begin{aligned}
\frac{d}{dt}(|\psi\rangle\langle\psi|) &= \frac{d|\psi\rangle}{dt}\langle\psi| + |\psi\rangle\frac{d\langle\psi|}{dt} \\
&= -\frac{i}{\hbar}\left(\hat{H} - i\alpha\left[\hat{H}, |\psi\rangle\langle\psi|\right]\right)|\psi\rangle\langle\psi| + \frac{i}{\hbar}|\psi\rangle\langle\psi|\left(\hat{H} + i\alpha\left[|\psi\rangle\langle\psi|, \hat{H}\right]\right) \\
&= \frac{i}{\hbar}\left[|\psi\rangle\langle\psi|, \hat{H}\right] - \frac{\alpha}{\hbar}\left[|\psi\rangle\langle\psi|, \left[|\psi\rangle\langle\psi|, \hat{H}\right]\right],
\end{aligned}$$

and finally with $\hat{\rho} = |\psi\rangle\langle\psi|$

$$\frac{d\hat{\rho}}{dt} = \frac{i}{\hbar}[\hat{\rho}, \hat{H}] - \frac{\alpha}{\hbar}[\hat{\rho}, [\hat{\rho}, \hat{H}]]. \quad (7)$$

III. DERIVATION OF THE LANDAU-LIFSHITZ-BLOCH EQ. STARTING FROM THE VON NEUMANN EQ. USING GEOMETRIC ALGEBRA

Starting point is the following von Neumann equation:

$$\frac{d\hat{\rho}}{dt} = \frac{i}{\hbar}[\hat{\rho}, \hat{H}] - \frac{\alpha_{\text{tr}}}{\hbar}[\hat{\rho}, [\hat{\rho}, \hat{H}]] + 2\frac{\alpha_1}{\hbar}(\hat{\rho}\hat{H})\hat{\rho}, \quad (8)$$

with the assumption that the density operator $\hat{\rho}$ is given by:

$$\hat{\rho} = \frac{1}{2}(\hat{\mathbf{1}} + \langle\hat{\sigma}\rangle \cdot \hat{\sigma}), \quad (9)$$

Furthermore, we assume that the Hamiltonian can be written as:

$$\hat{H} = -\frac{g\mu_B}{2}\mathbf{B} \cdot \hat{\sigma}, \quad (10)$$

with g the g-factor and μ_B Bohr magneton for the correct dimension. $\hat{\sigma} = (\hat{\sigma}_x, \hat{\sigma}_y, \hat{\sigma}_z)$ is the Pauli vector with $\hat{\sigma}_\eta$, $\eta \in \{x, y, z\}$ the Pauli Matrices and $\hat{\mathbf{1}}$ the identity matrix:

$$\hat{\sigma}_x = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}, \quad \hat{\sigma}_y = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix}, \quad \hat{\sigma}_z = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}, \quad \hat{\mathbf{1}} = \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}. \quad (11)$$

A view words about Eq. (9) and (10): As in the Euclidean space where we are able to express any vector in \mathbb{R}^3 as a linear combination of the basis vectors $\hat{\mathbf{x}}$, $\hat{\mathbf{y}}$, and $\hat{\mathbf{z}}$: $\mathbf{r} = x\hat{\mathbf{x}} + y\hat{\mathbf{y}} + z\hat{\mathbf{z}}$, we can (as an isomorphism) do the same with the Pauli matrices e.g.:

$$\mathcal{P} = \mathbf{P} \cdot \hat{\sigma} = P_x\hat{\sigma}_x + P_y\hat{\sigma}_y + P_z\hat{\sigma}_z. \quad (12)$$

Here, we have set $\mathbf{P} = \langle\hat{\sigma}\rangle$. Or, alternatively written as matrix:

$$\mathcal{P} = \begin{pmatrix} P_z & P_x - iP_y \\ P_x + iP_y & -P_z \end{pmatrix}. \quad (13)$$

$\mathcal{B} = -\mathbf{B} \cdot \hat{\sigma}$ is defined in a similar way.

Lets focus on the von Neumann equation. With aid of (9) the left hand side of Eq. (8) can be written as:

$$\frac{d\hat{\rho}}{dt} = \frac{d\mathbf{P}}{dt} \cdot \frac{\hat{\sigma}}{2} . \quad (14)$$

Please notice, here we are in the Schrödinger picture, meaning that the operators are time independent.

The precessional term (first term) of Eq. (8) contains the following commutator:

$$[\hat{\rho}, \hat{H}] = \frac{g\mu_B}{4} [\mathcal{P}, \mathcal{B}] = \frac{g\mu_B}{2} \mathcal{P} \wedge \mathcal{B} . \quad (15)$$

The wedge product in Eq. (15) is defined as:

$$\mathcal{P} \wedge \mathcal{B} = iP^n B^m \epsilon_{nml} \hat{\sigma}_l , \quad (16)$$

where ϵ_{nml} is the Levi-Civita tensor. Here, the Einstein sum convention has been used. Comparison with the vector product:

$$\mathbf{a} \times \mathbf{b} = a^n b^m \epsilon_{nml} \hat{\mathbf{e}}_l , \quad (17)$$

where $\hat{\mathbf{e}}_l$ is a unit vector perpendicular to \mathbf{a} and \mathbf{b} , leads to

$$\mathcal{P} \wedge \mathcal{B} = i(\mathbf{P} \times \mathbf{B})^l \hat{\sigma}_l = i(\mathbf{P} \times \mathbf{B}) \cdot \hat{\sigma} . \quad (18)$$

Next term is the transversal relaxation term which contains the double commutator:

$$[\hat{\rho}, [\hat{\rho}, \hat{H}]] = \frac{g\mu_B}{8} [\mathcal{P}, [\mathcal{P}, \mathcal{B}]] . \quad (19)$$

The inner commutator has been already calculated [see Eq. (15) and following equations].

The result can be written as:

$$[\hat{\rho}, \hat{H}] = \frac{g\mu_B}{4} [\mathcal{P}, \mathcal{B}] = \frac{ig\mu_B}{2} (\mathbf{P} \times \mathbf{B}) \cdot \hat{\sigma} = \frac{ig\mu_B}{2} \mathbf{A} \cdot \hat{\sigma} = \frac{ig\mu_B}{2} \mathcal{A} , \quad (20)$$

therefore:

$$[\hat{\rho}, [\hat{\rho}, \hat{H}]] = \frac{ig\mu_B}{4} [\mathcal{P}, \mathcal{A}] = \frac{i^2 g\mu_B}{2} (\mathbf{P} \times \mathbf{A}) \cdot \hat{\sigma} = -g\mu_B (\mathbf{P} \times (\mathbf{P} \times \mathbf{B})) \cdot \frac{\hat{\sigma}}{2} . \quad (21)$$

With this results and the gyromagnetic ratio $\gamma = g\mu_B/\hbar$, the von Neumann equation without longitudinal relaxation term:

$$\frac{d\hat{\rho}}{dt} = \frac{i}{\hbar} [\hat{\rho}, \hat{H}] - \frac{\alpha_{tr}}{\hbar} [\hat{\rho}, [\hat{\rho}, \hat{H}]] , \quad (22)$$

is given as:

$$\frac{d\mathbf{P}}{dt} = \gamma \mathbf{P} \times \mathbf{B} - \alpha_{\text{tr}} \gamma (\mathbf{P} \times (\mathbf{P} \times \mathbf{B})) . \quad (23)$$

Here, the $\cdot \hat{\sigma}/2$ term on both sides has been already skipped.

The results above can be proved by the direct multiplication and subtraction of the matrices. This shall be shown only for the precessional term, the transversal relaxation term is similar. For the commutator $[\hat{\rho}, \hat{H}] = -\frac{\gamma}{2}[\mathcal{P}, \mathcal{B}] = -\frac{\gamma}{4}(\mathcal{P}\mathcal{B} - \mathcal{B}\mathcal{P})$ we have:

$$\mathcal{P}\mathcal{B} = \begin{pmatrix} P_z B_z + (P_x - iP_y)(B_x + iB_y) & P_z(B_x - iB_y) - B_z(P_x - iP_y) \\ B_z(P_x + iP_y) - P_z(B_x + iB_y) & P_z B_z + (P_x + iP_y)(B_x - iB_y) \end{pmatrix} \quad (24)$$

and

$$\mathcal{B}\mathcal{P} = \begin{pmatrix} B_z P_z + (B_x - iB_y)(P_x + iP_y) & B_z(P_x - iP_y) - P_z(B_x - iB_y) \\ P_z(B_x + iB_y) - B_z(P_x + iP_y) & B_z P_z + (B_x + iB_y)(P_x - iP_y) \end{pmatrix} \quad (25)$$

and therefore:

$$\begin{aligned} \mathcal{P}\mathcal{B} - \mathcal{B}\mathcal{P} &= 2i \begin{pmatrix} (P_x B_y - P_y B_x) & (P_y B_z - P_z B_y) - i(P_z B_x - P_x B_z) \\ (P_y B_z - P_z B_y) + i(P_z B_x - P_x B_z) & -(P_x B_y - P_y B_x) \end{pmatrix} \\ &= 2i \begin{pmatrix} (\mathbf{P} \times \mathbf{B})_z & (\mathbf{P} \times \mathbf{B})_x - i(\mathbf{P} \times \mathbf{B})_y \\ (\mathbf{P} \times \mathbf{B})_x + i(\mathbf{P} \times \mathbf{B})_y & -(\mathbf{P} \times \mathbf{B})_z \end{pmatrix} . \end{aligned} \quad (26)$$

Finally with:

$$[\mathcal{P}, \mathcal{B}] = \mathcal{P}\mathcal{B} - \mathcal{B}\mathcal{P} = 2i(\mathbf{P} \times \mathbf{B}) \cdot \hat{\sigma} , \quad (27)$$

we find:

$$\frac{i}{\hbar}[\hat{\rho}, \hat{H}] = -\frac{i\gamma}{4}[\mathcal{P}, \mathcal{B}] = \gamma(\mathbf{P} \times \mathbf{B}) \cdot \frac{\hat{\sigma}}{2} . \quad (28)$$

In the case of the longitudinal relaxation we have the following term:

$$\frac{2\alpha_1}{\hbar}(\hat{\rho}\hat{H})\hat{\rho} = -\frac{2\alpha_1\gamma}{4\hbar}((\mathbf{P} \cdot \hat{\sigma})(\mathbf{B} \cdot \hat{\sigma})) \left(\mathbf{P} \cdot \frac{\hat{\sigma}}{2} \right) = -\frac{\alpha_1\gamma}{2}\langle \mathcal{P}, \mathcal{B} \rangle \cdot \left(\mathbf{P} \cdot \frac{\hat{\sigma}}{2} \right) . \quad (29)$$

As in the classical LLB equation where we have with $(\mathbf{m} \cdot \mathbf{B})\mathbf{m}$ a scalar product (inner product of two vectors in the Euclidian space) we have to deal here with the inner product between the matrices \mathcal{P} and \mathcal{B} :

$$\langle \mathcal{P}, \mathcal{B} \rangle = \text{Tr}(\mathcal{P}\mathcal{B}^*) . \quad (30)$$

The star index “ \star ” means conjugate transposed. In our case, \mathcal{B} is Hermitian $\mathcal{B}^\star = \mathcal{B}$ and therefore:

$$\langle \mathcal{P}, \mathcal{B} \rangle = \text{Tr}(\mathcal{P}\mathcal{B}) . \quad (31)$$

With:

$$\text{Tr}(\mathcal{P}\mathcal{B}) = \text{Tr} \begin{pmatrix} P_z B_z + (P_x - iP_y)(B_x + iB_y) & P_z(B_x - iB_y) - B_z(P_x - iP_y) \\ B_z(P_x + iP_y) - P_z(B_x + iB_y) & P_z B_z + (P_x + iP_y)(B_x - iB_y) \end{pmatrix} = 2\mathbf{P} \cdot \mathbf{B} ,$$

we get finally:

$$\langle \mathcal{P}, \mathcal{B} \rangle = 2(\mathbf{P} \cdot \mathbf{B}) , \quad (32)$$

where $\mathbf{P} \cdot \mathbf{B}$ is the scalar product between two vectors in \mathbb{R}^3 and therefore a scalar.

Using this result we are able to write:

$$\frac{2\alpha_1}{\hbar}(\hat{\rho}\hat{H})\hat{\rho} = -\frac{\alpha_1\gamma}{2}\langle \mathcal{P}, \mathcal{B} \rangle \cdot \left(\mathbf{P} \cdot \frac{\hat{\sigma}}{2} \right) = -\alpha_1\gamma(\mathbf{P} \cdot \mathbf{B}) \cdot \left(\mathbf{P} \cdot \frac{\hat{\sigma}}{2} \right) , \quad (33)$$

and therefore the von Neumann Eq. (8) as:

$$\frac{d\mathbf{P}}{dt} = \gamma\mathbf{P} \times \mathbf{B} - \alpha_{\text{tr}}\gamma(\mathbf{P} \times (\mathbf{P} \times \mathbf{B})) - \alpha_1\gamma(\mathbf{P} \cdot \mathbf{B}) \cdot \mathbf{P} . \quad (34)$$

Again, the $\cdot\hat{\sigma}/2$ term has been skipped on both sides of this differential equation.

IV. ANALYTICAL PROOF USING THE HEISENBERG EQUATION

To derive the Heisenberg equation:

$$\frac{d\langle \hat{\mathbf{S}} \rangle}{dt} = -\frac{i}{\hbar}\langle [\hat{\mathbf{S}}, \hat{H}] \rangle - \frac{\alpha_{\text{tr}}}{\hbar}\langle \{ \hat{\mathbf{S}}, \hat{H} \} \rangle + 2\frac{\alpha_{\text{tr}} + \alpha_1}{\hbar}\langle \hat{H} \rangle \langle \hat{\mathbf{S}} \rangle , \quad (35)$$

we can start from the von Neumann Eq. (8) and write this equation in the alternative form:

$$[\hat{\rho}, [\hat{\rho}, \hat{H}]] = \{ \hat{\rho}, \hat{H} \} - 2\hat{\rho}\hat{H}\hat{\rho} , \quad (36)$$

$$\frac{d\hat{\rho}}{dt} = \frac{i}{\hbar}[\hat{\rho}, \hat{H}] - \frac{\alpha_{\text{tr}}}{\hbar}\{ \hat{\rho}, \hat{H} \} + 2\frac{\alpha_{\text{tr}} + \alpha_1}{\hbar}(\hat{\rho}\hat{H})\hat{\rho} . \quad (37)$$

After adding Tr and $\hat{\mathbf{S}}$ to calculate the spin expectation values $\langle \hat{\mathbf{S}} \rangle$ we end up with the Heisenberg Eq. (35). It is convenient to normalize the spin expectation values $\langle \hat{\mathbf{S}} \rangle$ (dividing both sides of Eq. (35) by $\hbar S$). In the case of $S = 1/2$ this leads to:

$$\frac{d\langle \hat{\sigma} \rangle}{dt} = -\frac{i}{\hbar}\langle [\hat{\sigma}, \hat{H}] \rangle - \frac{\alpha_{\text{tr}}}{\hbar}\langle \{ \hat{\sigma}, \hat{H} \} \rangle + 2\frac{\alpha_{\text{tr}} + \alpha_1}{\hbar}\langle \hat{H} \rangle \langle \hat{\sigma} \rangle . \quad (38)$$

At this point we have to say that we have to add an additional $\langle\psi|\psi\rangle$ to the second term to be conform with the time dependent Schrödinger equation given in the paper. We will see that this term is also needed here. Then, with $\tilde{\alpha}_{\text{tr}} = \alpha_{\text{tr}}\langle\psi|\psi\rangle$ the final Heisenberg equation is given by:

$$\frac{d\langle\hat{\sigma}\rangle}{dt} = -\frac{i}{\hbar}\langle[\hat{\sigma}, \hat{H}]\rangle - \frac{\tilde{\alpha}_{\text{tr}}}{\hbar}\langle\{\hat{\sigma}, \hat{H}\}\rangle + 2\frac{\alpha_{\text{tr}} + \alpha_l}{\hbar}\langle\hat{H}\rangle\langle\hat{\sigma}\rangle. \quad (39)$$

In the following we assume that the Hamiltonian \hat{H} is given by:

$$\hat{H} = -\frac{g\mu_B}{2}\mathbf{B} \cdot \hat{\sigma}. \quad (40)$$

Now, lets look for the terms on the right hand side and take into account that $\gamma = g\mu_B/\hbar$:

The first term describes the precessional motion:

$$-\frac{i}{\hbar}\langle[\hat{\sigma}, \hat{H}]\rangle = \frac{i\gamma}{2}\langle[\hat{\sigma}, \mathbf{B} \cdot \hat{\sigma}]\rangle. \quad (41)$$

This is an vector, therefore it is more convenient to look for the components $u, v, w \in \{x, y, z\}$ separately:

$$\frac{i\gamma}{2}B^v\langle[\hat{\sigma}_u, \hat{\sigma}_v]\rangle = \frac{2i^2}{2}\gamma B^v\epsilon_{uvw}\langle\hat{\sigma}_w\rangle = \gamma(\mathbf{B} \times \langle\hat{\sigma}\rangle)_u. \quad (42)$$

Here, we have used the commutator relation $[\hat{\sigma}_u, \hat{\sigma}_v] = 2i\epsilon_{uvw}\hat{\sigma}_w$ and the Einstein sum convention after which we have to sum over identical indices. We find finally, written as vector :

$$-\frac{i}{\hbar}\langle[\hat{\sigma}, \hat{H}]\rangle = \gamma\mathbf{B} \times \langle\hat{\sigma}\rangle. \quad (43)$$

The second term is:

$$-\frac{\tilde{\alpha}_{\text{tr}}}{\hbar}\langle\{\hat{\sigma}, \hat{H}\}\rangle = \frac{\gamma\alpha_{\text{tr}}}{2}\langle\{\hat{\sigma}, \mathbf{B} \cdot \hat{\sigma}\}\rangle\langle\psi|\psi\rangle. \quad (44)$$

Again, this is a vector and it is more convenient to look for one component:

$$\frac{\gamma\alpha_{\text{tr}}}{2}B^v\langle\{\hat{\sigma}_u, \hat{\sigma}_v\}\rangle\langle\psi|\psi\rangle = \gamma\alpha_{\text{tr}}B_u[\langle\psi|\psi\rangle]^2. \quad (45)$$

Here, we have used the anti-commutator relation $\{\hat{\sigma}_u, \hat{\sigma}_v\} = 2\delta_{uv}\hat{\mathbf{1}}$ and $\langle\hat{\mathbf{1}}\rangle = \langle\psi|\hat{\mathbf{1}}|\psi\rangle = \langle\psi|\psi\rangle$. Written as vector:

$$-\frac{\tilde{\alpha}_{\text{tr}}}{\hbar}\langle\{\hat{\sigma}, \hat{H}\}\rangle = \gamma\alpha_{\text{tr}}\mathbf{B}[\langle\psi|\psi\rangle]^2. \quad (46)$$

Introducing the vector $\tilde{\mathbf{m}}$ with $\tilde{\mathbf{m}}^2 = \tilde{\mathbf{m}} \cdot \tilde{\mathbf{m}} = 1$ and the relation $\tilde{\mathbf{m}}\langle\psi|\psi\rangle = \langle\hat{\sigma}\rangle$, and $\langle\hat{\sigma}\rangle^2 = \langle\hat{\sigma}\rangle \cdot \langle\hat{\sigma}\rangle = [\langle\psi|\psi\rangle]^2 \leq 1$ we get:

$$-\frac{\tilde{\alpha}_{\text{tr}}}{\hbar}\langle\{\hat{\sigma}, \hat{H}\}\rangle = \gamma\alpha_{\text{tr}}\mathbf{B}\tilde{\mathbf{m}}^2[\langle\psi|\psi\rangle]^2 = \gamma\alpha_{\text{tr}}\mathbf{B}(\langle\hat{\sigma}\rangle \cdot \langle\hat{\sigma}\rangle). \quad (47)$$

In the case $\alpha_1 = 0$ (no longitudinal relaxation) we have $\langle\psi|\psi\rangle = 1$ and therefore $\langle\hat{\sigma}\rangle^2 = 1$. However, this is not the case for $\alpha_1 \neq 0$. Here, we have $0 \leq \langle\psi|\psi\rangle \leq 1$. Therefore, during the calculation it was needed to use $[\langle\psi|\psi\rangle]^2$: one $\langle\psi|\psi\rangle$ came from the expectation value of the anti-commutator $\langle\{\hat{\sigma}_u, \hat{\sigma}_v\}\rangle = 2\delta_{uv}\langle\psi|\psi\rangle$ and the second from the modification: $\tilde{\alpha}_{\text{tr}} = \alpha_{\text{tr}}\langle\psi|\psi\rangle$. This means that the modification we have introduced in the paper is also needed here to get the correct result in the case $\alpha_1 \neq 0$.

The last term is given by:

$$2\frac{\alpha_{\text{tr}} + \alpha_1}{\hbar}\langle\hat{H}\rangle\langle\hat{\sigma}\rangle = -\gamma(\alpha_{\text{tr}} + \alpha_1)(\mathbf{B} \cdot \langle\hat{\sigma}\rangle)\langle\hat{\sigma}\rangle. \quad (48)$$

If we combine this (only the transversal relaxation, meaning only the part with α_{tr}) with the second term we find:

$$\gamma\alpha_{\text{tr}}\left(\mathbf{B}(\langle\hat{\sigma}\rangle \cdot \langle\hat{\sigma}\rangle) - (\mathbf{B} \cdot \langle\hat{\sigma}\rangle)\langle\hat{\sigma}\rangle\right) = -\gamma\alpha_{\text{tr}}\langle\hat{\sigma}\rangle \times \left(\langle\hat{\sigma}\rangle \times \mathbf{B}\right), \quad (49)$$

and in total:

$$\frac{d\langle\hat{\sigma}\rangle}{dt} = \gamma\mathbf{B} \times \langle\hat{\sigma}\rangle - \gamma\alpha_{\text{tr}}\langle\hat{\sigma}\rangle \times \left(\langle\hat{\sigma}\rangle \times \mathbf{B}\right) - \gamma\alpha_1(\mathbf{B} \cdot \langle\hat{\sigma}\rangle)\langle\hat{\sigma}\rangle. \quad (50)$$

V. SPIN DENSITY OPERATOR FOR $S = 1/2$

The wave function of a single spin $S = 1/2$ is given by:

$$|\psi\rangle = \cos\frac{\theta}{2}|\uparrow\rangle + \sin\frac{\theta}{2}e^{i\phi}|\downarrow\rangle. \quad (51)$$

The same is true for the $S = 1/2$ coherent spin state:

$$\begin{aligned} |\psi\rangle &= e^{-\frac{i\phi\hat{S}_z}{\hbar}}e^{-\frac{i\theta\hat{S}_y}{\hbar}}|\uparrow\rangle \\ &= \cos\frac{\theta}{2}|\uparrow\rangle + \sin\frac{\theta}{2}e^{i\phi}|\downarrow\rangle. \end{aligned} \quad (52)$$

Then, the corresponding density operator is given by:

$$\begin{aligned} \hat{\rho} = |\psi\rangle\langle\psi| &= \left(\cos\frac{\theta}{2}|\uparrow\rangle + \sin\frac{\theta}{2}e^{i\phi}|\downarrow\rangle\right)\left(\langle\uparrow|\cos\frac{\theta}{2} + \langle\downarrow|\sin\frac{\theta}{2}e^{-i\phi}\right) \\ &= \cos^2\frac{\theta}{2}|\uparrow\rangle\langle\uparrow| + \cos\frac{\theta}{2}\sin\frac{\theta}{2}e^{-i\phi}|\uparrow\rangle\langle\downarrow| + \cos\frac{\theta}{2}\sin\frac{\theta}{2}e^{i\phi}|\downarrow\rangle\langle\uparrow| + \sin^2\frac{\theta}{2}|\downarrow\rangle\langle\downarrow|. \end{aligned} \quad (53)$$

With $\sin(2\theta) = 2 \sin \theta \cos \theta$, $\sin^2 \theta = 1/2 - 1/2 \cos(2\theta)$ and $\cos^2 \theta = 1/2 + 1/2 \cos(2\theta)$, we are able to write:

$$\hat{\rho} = |\psi\rangle\langle\psi| = \frac{1}{2} \left((1 + \cos \theta) |\uparrow\rangle\langle\uparrow| + \sin \theta e^{-i\phi} |\uparrow\rangle\langle\downarrow| + \sin \theta e^{i\phi} |\downarrow\rangle\langle\uparrow| + (1 - \cos \theta) |\downarrow\rangle\langle\downarrow| \right). \quad (54)$$

With $|\uparrow\rangle = \begin{pmatrix} 1 \\ 0 \end{pmatrix}$, and $|\downarrow\rangle = \begin{pmatrix} 0 \\ 1 \end{pmatrix}$ we can write this as matrix:

$$\hat{\rho} = \frac{1}{2} \begin{pmatrix} 1 + \cos \theta & \sin \theta e^{-i\phi} \\ \sin \theta e^{i\phi} & 1 - \cos \theta \end{pmatrix}. \quad (55)$$

With $\exp(\pm i\phi) = \cos \phi \pm i \sin \phi$, $\langle \hat{\sigma}_x \rangle = \sin \theta \cos \phi$, $\langle \hat{\sigma}_y \rangle = \sin \theta \sin \phi$, and $\langle \hat{\sigma}_z \rangle = \cos \theta$ we get finally:

$$\hat{\rho} = \frac{1}{2} \begin{pmatrix} 1 + \langle \hat{\sigma}_z \rangle & \langle \hat{\sigma}_x \rangle - i \langle \hat{\sigma}_y \rangle \\ \langle \hat{\sigma}_x \rangle + i \langle \hat{\sigma}_y \rangle & 1 - \langle \hat{\sigma}_z \rangle \end{pmatrix}. \quad (56)$$

This can be written in a more compact form as:

$$\hat{\rho} = \frac{1}{2} (\hat{\mathbf{1}} + \langle \hat{\sigma} \rangle \cdot \hat{\sigma}), \quad (57)$$

with the Pauli matrices $\hat{\sigma}_\eta$, $\eta \in \{x, y, z\}$, and $\hat{\mathbf{1}}$ the identity matrix.

VI. DERIVATION OF THE QLLB EQ. USING DENSITY OPERATOR FOR AN ARBITRARY SPIN QUANTUM NUMBER S

The density operator for any spin quantum number S can be written as [1]:

$$\hat{\rho} = \frac{1}{2S+1} \hat{\mathbf{1}} + \mathbf{m} \cdot \hat{\mathbf{S}} + \text{higher order tensors}, \quad (58)$$

with

$$\mathbf{m} = \frac{\langle \hat{\mathbf{S}} \rangle}{\hbar S} = \mathbf{e}_S, \quad (59)$$

where \mathbf{e}_S is a normalized vector:

$$\mathbf{e}_S = \begin{pmatrix} \cos \phi \sin \theta \\ \sin \phi \sin \theta \\ \cos \theta \end{pmatrix}. \quad (60)$$

The number and appearance of the higher order tensor terms is given by the spin quantum number S : Such terms appear only for $S > 1/2$ [2, 3]. Furthermore, the number of higher order tensor terms increases with increasing S . For the conventional spin dynamics the vector terms [second term in Eq. (58)] are important. The higher order tensor terms will lead to separate differential equations which don't interfere with the dynamics of the Bloch vector \mathbf{m} .

Now, the quantum Landau-Lifshitz-Bloch (qLLB) equation is defined as:

$$i\hbar \frac{d}{dt} |\psi\rangle = \left(\hat{H} - i\alpha_{\text{tr}}[\langle\psi|\psi\rangle \hat{H} - \langle\hat{H}\rangle] + i\alpha_1 \langle\hat{H}\rangle \right) |\psi\rangle , \quad (61)$$

and the corresponding von Neumann equation is:

$$\frac{d\hat{\rho}}{dt} = \frac{i}{\hbar} [\hat{\rho}, \hat{H}] - \frac{\alpha_{\text{tr}}}{\hbar} [\hat{\rho}, [\hat{\rho}, \hat{H}]] + \frac{\alpha_1}{\hbar} (\hat{\rho} \hat{H}) \hat{\rho} . \quad (62)$$

In both differential equations (qLLB and von Neumann equation) the terms on the right hand side correspond to a precessional motion (first term), transversal (second term) and longitudinal relaxation (last term). In the von Neumann equation within the publication the longitudinal relaxation term is defined with an additional factor two which has been introduced to make a symmetric decoupling going from the von Neumann equation to the TDSE. However, this term only scales the strength of the relaxation and can be skipped because the function α_1 has to be defined anyhow. Furthermore, the decoupling of the von Neumann equation to derive the TDSE can be also asymmetric meaning that only the one differential equation contains the longitudinal relaxation term which becomes after skipping $\langle\psi|$ the searched TDSE while the corresponding conjugate complex TDSE does not contain this term.

In the following we have assumed that the Hamilton operator \hat{H} is given by:

$$\hat{H} = -\gamma \mathbf{B} \cdot \hat{\mathbf{S}} , \quad (63)$$

with \mathbf{B} an effective field.

Inserting this equation into Eq. (62) together with Eq. (58) and (63) we find the following differential equation:

$$\frac{\partial \mathbf{m}}{\partial t} = \gamma \mathbf{m} \times \mathbf{B} - \gamma \tilde{\alpha}_{\text{tr}} \mathbf{m} \times (\mathbf{m} \times \mathbf{B}) - \gamma \tilde{\alpha}_1 (\mathbf{m} \cdot \mathbf{B}) \mathbf{m} . \quad (64)$$

In detail: For the left hand side we immediately find:

$$\frac{\partial \hat{\rho}}{\partial t} = \frac{\partial \mathbf{m}}{\partial t} \cdot \hat{\mathbf{S}} . \quad (65)$$

Now, from the connection between classical physics and quantum mechanics we know that the commutator $[\hat{A}, \hat{B}]$ and the Poisson bracket $\{A, B\}$ are connected by $[\hat{A}, \hat{B}] \leftrightarrow i\hbar\{A, B\}$. Then, the Poisson bracket of a classical spin system is given by [4, 5]:

$$\{A, B\} = \frac{\partial A}{\partial S_n} \frac{\partial B}{\partial S_m} S_l \epsilon_{nml}, \quad (66)$$

where the Einstein sum convention has been used. A , B and C are functions of a spin tensor \mathbf{S} of rank n and S_n, S_m, \dots, S_v are the components of this spin tensor. As mentioned before we are just interested in the spin operator $\hat{\mathbf{S}}$ and therefore in spin tensors \hat{S}_n of first rank. This means: S_n, S_m, \dots, S_v are the spin components S_x, S_y , and S_z . Furthermore, ϵ_{nml} is the Levi-Civita tensor. With this informations it is easy to calculate the commutator $[\hat{\rho}, \hat{H}]$:

$$\begin{aligned} \frac{i}{\hbar}[\hat{\rho}, \hat{H}] &= -\frac{i\gamma}{\hbar}[\mathbf{m} \cdot \hat{\mathbf{S}}, \mathbf{B} \cdot \hat{\mathbf{S}}] = -i\hbar \frac{i}{\hbar} \gamma \{\mathbf{m} \cdot \hat{\mathbf{S}}, \mathbf{B} \cdot \hat{\mathbf{S}}\} = \gamma \{\mathbf{m} \cdot \hat{\mathbf{S}}, \mathbf{B} \cdot \hat{\mathbf{S}}\} \\ &= \gamma m_u B_v \frac{\partial \hat{S}_u}{\partial \hat{S}_n} \frac{\partial \hat{S}_v}{\partial \hat{S}_m} \hat{S}_l \epsilon_{nml} = \gamma m_u B_v \hat{S}_l \delta_{un} \delta_{vm} \epsilon_{nml} = \gamma (\mathbf{m} \times \mathbf{B}) \cdot \hat{\mathbf{S}}. \end{aligned} \quad (67)$$

This result can be easily proved with the commutator:

$$[\hat{S}_u, \hat{S}_v] = i\hbar \epsilon_{uvw} \hat{S}_w. \quad (68)$$

The commutator $[\hat{\rho}, \hat{H}]$ can be written as:

$$\frac{i}{\hbar}[\hat{\rho}, \hat{H}] = -\frac{i\gamma}{\hbar}[\mathbf{m} \cdot \hat{\mathbf{S}}, \mathbf{B} \cdot \hat{\mathbf{S}}] = -\frac{i\gamma}{\hbar} m_u B_v [\hat{S}_u, \hat{S}_v] = \gamma m_u B_v \hat{S}_w \epsilon_{uvw} = \gamma (\mathbf{m} \times \mathbf{B}) \cdot \hat{\mathbf{S}}, \quad (69)$$

where with the definition of the vector product via Levi-Civita tensor it is easy to reproduce the above result without using the Poisson bracket.

The next term is the transversal relaxation term. Here, we have the double commutator:

$$\frac{\alpha_{\text{tr}}}{\hbar}[\hat{\rho}, [\hat{\rho}, \hat{H}]] = -i\alpha_{\text{tr}}\gamma[\mathbf{m} \cdot \hat{\mathbf{S}}, \mathbf{v} \cdot \hat{\mathbf{S}}]. \quad (70)$$

Here, we have used the fact that we have already solved the commutator:

$$[\hat{\rho}, \hat{H}] = -i\hbar\gamma (\mathbf{m} \times \mathbf{B}) \cdot \hat{\mathbf{S}} = -i\hbar\gamma \mathbf{v} \cdot \hat{\mathbf{S}}. \quad (71)$$

Then the corresponding commutator with spin operators $\hat{\mathbf{S}}$ is given by :

$$-i\alpha_{\text{tr}}\gamma[\mathbf{m} \cdot \hat{\mathbf{S}}, \mathbf{v} \cdot \hat{\mathbf{S}}] = \alpha_{\text{tr}}\gamma\hbar (\mathbf{m} \times \mathbf{v}) \cdot \hat{\mathbf{S}} = \alpha_{\text{tr}}\gamma\hbar (\mathbf{m} \times (\mathbf{m} \times \mathbf{B})) \cdot \hat{\mathbf{S}}, \quad (72)$$

and therefore:

$$\frac{\alpha_{\text{tr}}}{\hbar}[\hat{\rho}, [\hat{\rho}, \hat{H}]] = \alpha_{\text{tr}}\hbar(\mathbf{m} \times (\mathbf{m} \times \mathbf{B})) \cdot \hat{\mathbf{S}}. \quad (73)$$

Finally, the longitudinal relaxation term is given by:

$$\frac{\alpha_1}{\hbar}(\hat{\rho}\hat{H})\hat{\rho} = -\frac{\alpha_1\gamma}{\hbar} \left((\mathbf{m} \cdot \hat{\mathbf{S}})(\mathbf{B} \cdot \hat{\mathbf{S}}) \right) \mathbf{m} \cdot \hat{\mathbf{S}}. \quad (74)$$

Here, we have already skipped the noncontributing term with the identity operator $\hat{\mathbf{1}}$. With $\mathcal{M} = \mathbf{m} \cdot \hat{\mathbf{S}}$, $\mathcal{B} = \mathbf{B} \cdot \hat{\mathbf{S}}$, and the definition of the inner product using the trace we have:

$$\frac{\alpha_1}{\hbar}(\hat{\rho}\hat{H})\hat{\rho} = -\frac{\alpha_1\gamma}{\hbar} \langle \mathcal{M}, \mathcal{B} \rangle \mathbf{m} \cdot \hat{\mathbf{S}} = -\frac{\alpha_1\gamma}{N\hbar} \text{Tr}(\mathcal{M}\mathcal{B}) \mathbf{m} \cdot \hat{\mathbf{S}}, \quad (75)$$

where $N = 2S + 1$ is the normalization because the inner product does not necessarily deliver normalized results. In the case $S = 1/2$ the normalization was not needed due to the fact that there the additional $1/2$ factors corresponding the transformation $\hat{\mathbf{S}} = (\hbar/2)\hat{\sigma}$ have done the job and the inner product was normalized. In general this is not the case and therefore a normalization needed. It seems that the additional factor two in front of the longitudinal relaxation term works fine with $S = 1/2$ but not with $S > 1/2$.

Now, with aid of the geometric product (Grassmann):

$$\begin{aligned} \mathcal{M}\mathcal{B} &= \mathcal{M} \cdot \mathcal{B} + \mathcal{M} \wedge \mathcal{B} \\ &= (\mathbf{m} \cdot \mathbf{B}) \hat{\mathbf{1}} + i(\mathbf{m} \times \mathbf{B}) \cdot \hat{\mathbf{S}}, \end{aligned} \quad (76)$$

it is easy to calculate the trace using:

$$\text{Tr}(\hat{\mathbf{1}}) = 2S + 1 \quad \text{and} \quad \text{Tr}(\hat{\mathbf{S}}) = 0. \quad (77)$$

Therefore the trace is given by:

$$\frac{1}{2S+1} \text{Tr}(\mathcal{M}\mathcal{B}) = \frac{1}{2S+1} \left[(\mathbf{m} \cdot \mathbf{B}) \text{Tr}(\hat{\mathbf{1}}) + i(\mathbf{m} \times \mathbf{B})_n \text{Tr}(\hat{S}_n) \right] = \mathbf{m} \cdot \mathbf{B}. \quad (78)$$

Finally, we find:

$$\frac{\alpha_1}{\hbar}(\hat{\rho}\hat{H})\hat{\rho} = -\frac{\alpha_1\gamma}{\hbar} (\mathbf{m} \cdot \mathbf{B}) \mathbf{m} \cdot \hat{\mathbf{S}}. \quad (79)$$

Bringing all parts together and skipping the $\hat{\mathbf{S}}$ on the right hand side in any term and setting $\alpha_{\text{tr}}\hbar = \tilde{\alpha}_{\text{tr}}$ as well $\alpha_1/\hbar = \tilde{\alpha}_1$ we get Eq. (64) as final result. Please notice that both

$\tilde{\alpha}_{\text{tr}}$ as well as $\tilde{\alpha}_l$ have the dimension of \hbar . This is similar to the Landau-Lifshitz constant λ which has the same dimension as the gyromagnetic ratio γ .

At the end a view words more about the inner product. As mentioned before in Euclidian vector space we can write any vector \mathbf{a} as a $n \times 1$ or $1 \times n$ matrix e.g. in \mathbb{R}^3 as $\mathbf{a} = (a_x, a_y, a_z)^T$. Alternative we can develop the vector with respect to its basis vectors $\hat{\mathbf{x}} = (1, 0, 0)^T$, $\hat{\mathbf{y}} = (0, 1, 0)^T$, and $\hat{\mathbf{z}} = (0, 0, 1)^T$: $\mathbf{a} = a_x \hat{\mathbf{x}} + a_y \hat{\mathbf{y}} + a_z \hat{\mathbf{z}}$. In the case of the von Neumann equation we are dealing with matrices $\mathcal{M} = \mathbf{m} \cdot \hat{\mathbf{S}}$, and $\mathcal{B} = \mathbf{B} \cdot \hat{\mathbf{S}}$ which can be also understood as vectors. Here the spin operators $\hat{S}_x, \hat{S}_y, \hat{S}_z$ have the same function as the basis vectors $\hat{\mathbf{x}}, \hat{\mathbf{y}}$, and $\hat{\mathbf{z}}$: $\mathcal{M} = m_x \hat{S}_x + m_y \hat{S}_y + m_z \hat{S}_z$, and $\mathcal{B} = B_x \hat{S}_x + B_y \hat{S}_y + B_z \hat{S}_z$. Similar to the Euclidian vector we can also write this as a $n \times 1$ or $1 \times n$ matrix: e.g. $\mathcal{M} = (m_x, m_y, m_z)^T$, and $\mathcal{B} = (B_x, B_y, B_z)^T$. With this we can write:

$$\mathcal{M}\mathcal{B} = (\mathbf{m} \cdot \hat{\mathbf{S}})(\mathbf{B} \cdot \hat{\mathbf{S}}) = (m_x, m_y, m_z) \begin{pmatrix} B_x \\ B_y \\ B_z \end{pmatrix} = \mathbf{m} \cdot \mathbf{B} , \quad (80)$$

and therefore:

$$\frac{\alpha_l}{\hbar}(\hat{\rho}\hat{H})\hat{\rho} = -\frac{\alpha_l\gamma}{\hbar}(\mathcal{M}\mathcal{B})\mathcal{M} = -\frac{\alpha_l\gamma}{\hbar}(\mathbf{m} \cdot \mathbf{B})\mathbf{m} \cdot \hat{\mathbf{S}} . \quad (81)$$

With this, it also becomes more clear why we need a normalization of the inner product using the trace: While the Euclidian basis vectors $\hat{\mathbf{x}}, \hat{\mathbf{y}}$, and $\hat{\mathbf{z}}$ are normalized it is not the case for the spin operator \hat{S}_x, \hat{S}_y , and \hat{S}_z . Moreover the size of this matrices change with changing spin quantum number S . The same is true for the corresponding identity matrix $\hat{\mathbf{1}}$. Therefore the norm $N = 2S + 1$ has to be adapted to S .

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